Optical-chemical-microphysical relationships and closure studies for mixed carbonaceous aerosols observed at Jeju Island; 3-laser photoacoustic spectrometer, particle sizing, and filter analysis

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Transport of aerosols in pollution plumes from the mainland Asian continent was observed in situ at Jeju, South Korea during the Cheju Asian Brown Cloud Plume-Asian Monsoon Experiment (CAPMEX) field campaign throughout August and September 2008 using a 3-laser photoacoustic spectrometer (PASS-3), chemical filter analysis, and size distributions. The PASS-3 directly measures the effects of morphology (e.g. coatings) on light absorption that traditional filter-based instruments are unable to address. Transport of mixed sulfate, carbonaceous, and nitrate aerosols from various Asian pollution plumes to Jeju accounted for 74% of the deployment days, showing large variations in their measured chemical and optical properties. Analysis of eight distinct episodes, spanning wide ranges of chemical composition, optical properties, and source regions, reveals that episodes with higher organic carbon (OC)/sulfate (SO_4^{2-}) and nitrate (NO_3^-)/SO_4^{2-} composition ratios exhibit lower single scatter albedo at shorter wavelengths (ω_{405}). We infer complex refractive indices (n–ik) as a function of wavelength for the high, intermediate, and low OC/SO_4^{2-} pollution episodes by using the observed particle size distributions and the measured optical properties. The smallest mean particle diameter corresponds to the high OC/SO_4^{2-} aerosol episode. The imaginary part of the refractive index (k) is greater for the high OC/SO_4^{2-} episode at all wavelengths. A distinct, sharp increase in k at short wavelength implies enhanced light absorption by OC, which accounts for 50% of the light absorption at 405 nm, in the high OC/SO_4^{2-} episode. Idealized analysis indicates increased absorption at 781 nm by factors greater than 3 relative to denuded black carbon in the laboratory. We hypothesize that coatings of black carbon cores are the mechanism of this enhancement. This implies that climate warming and atmospheric heating rates from black carbon particles can be significantly larger than have been estimated previously. The results of this study demonstrate ways in which atmospheric processing and mixing can amplify particle light absorption for carbonaceous aerosol, significantly at short wavelength, underscoring the need to understand and predict chemical composition effects on optical properties to accurately estimate the climate radiative forcing by mixed carbonaceous aerosols.

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